



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: :

John C. Parsons : Examiner: Peter Y. Choi

U.S. Serial No. 09/883,520 : Group Art Unit: 1771

Filed: June 18, 2001

Docket No. 1931.VIN

For: WATER DISPERSIBLE, SALT
SENSITIVE NONWOVEN MATERIALS

Mail Stop Amendment
Commissioner for Patents
P.O. Box 1450
Alexandria, Virginia 22313-1450

DECLARATION UNDER 37 CFR 1.132

Steven P. Pauls, co-inventor of the subject matter of the above-noted patent application hereby declares that:

1. He has worked in the field of polymer technology for 24 years, and that he is a co-inventor of the pending '520 application referenced above (sometimes referred to hereafter simply as the "present invention"), which is directed to fibrous webs having salt-sensitive latex binders.
2. That he understands from counsel that the claims have been rejected over United States Patent No. 5,631,317 to *Komatsu* and United States Patent No. 5,976,694 to *Tsai*, and that he is familiar with the references used in making those rejections.

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Additionally, he understands that the claims have been rejected as non-enabling with respect to the claimed glass transition temperature range.

3. That he makes this *Declaration* on personal knowledge of the facts stated herein.
4. The Present Invention relates to nonwoven webs which include a salt-sensitive latex polymer binder that comprises a polymer component which is emulsion polymerized in the presence of a colloid stabilizer. New claim 23 is reproduced below:

23. (New) A non-woven material comprising:
a) a web of fibers; and
b) a latex polymer binder applied to the web of fibers, wherein said latex polymer binder has a glass transition temperature of from -40°C to 105°C and comprises:
i) a polymer component which includes at least one hydrophilic monomer selected from the group consisting of acidic monomers containing a carboxylic acid moiety, dicarboxylic acid moiety, a sulfonic acid moiety, or combinations thereof; and at least one non-hydrophilic monomer selected from the group consisting of (meth) acrylates, maleates, (meth) acrylamides, vinyl esters, and combinations thereof,
and
ii) a polymeric colloid component,
wherein said polymer component is emulsion polymerized using said colloid component as a stabilizer, and
wherein said latex polymer composition forms films that are dispersible in tap water, and non-dispersible in aqueous solutions containing 0.5 weight percent or more of an inorganic salt.

The invention is thus directed to non-woven webs which retain their strength in the presence of a salt-containing medium, but readily disperse in tap water such that they may be flushed after use.

5. That, along with copending US Patent Application No. 09/540,033 (now US Patent No. 6,683,129 to *Eknoian*), the invention in the pending case represents a significant advancement in salt-sensitive technology because the salt-sensitive resins are made by emulsion polymerization. Emulsion polymerization techniques are described briefly as follows. Generally, in emulsion polymerization processes, a stabilizing

agent is provided to an aqueous medium where the stabilizing agent may include surfactants, emulsifiers, protective colloids or the like; the latex binders in the inventive compositions include at least a protective colloid as a stabilizer. The stabilizing agent has structure which typically includes both a hydrophilic portion and a hydrophobic portion such that the stabilizer orients in the reaction medium to form a plurality of “micelles” in the reaction medium which are generally spherical cell-like structures each having a size of from about 25 to 200 nm. When the monomer charge and initiator are added to the reaction medium, the polymerization proceeds within the micelles. The product latex that is produced by this process comprises a plurality of small polymer particles surrounded by the stabilizing agent, which are dispersed in the aqueous medium. Emulsion polymerization techniques are known in the art and are described in detail in the ‘033 application.

6. In contrast to the above noted emulsion polymerization techniques, the primary reference cited by the Examiner, ‘317 *Komatsu*, discloses solution polymerized polymers. Solution polymerization techniques are fundamentally different from emulsion polymerization, and generally involve dissolving the monomer components in an organic solvent and initiating the polymerization, where the reaction components and polymer product are dissolved in the organic solution. In solution polymerization processes, there is typically only one phase.
7. Specifically, the ‘317 *Komatsu* reference discloses salt-sensitive polymers that are polymerized by dissolving the monomers in a miscible acetone/water mixture, where after polymerization, the solvent is evaporated off such that the polymer is dispersed in the water component. Although the *Komatsu* patent refers to the polymer that is dispersed in water as an “emulsion” it is clear that the polymer is not emulsion polymerized, nor does it contain a colloid component or other stabilizers, both features that are embodied in the subject matter of the pending claims. The compositions in *Komatsu* are therefore not “emulsions” within the meaning of the pending application.

8. Additionally, the emulsion polymers produced according to the invention have a significantly different structure than solution polymers, even after the polymer composition coalesces into a film on a nonwoven web. Without being bound by a theory, it is believed the emulsion polymers form films by coalescence of discrete polymer particles which are separated by the stabilizing agent; this results in relatively weak bonds. Weaker bonds are advantageous in applications where water dispersibility is required. With solution polymerized resins, the polymer chains mix and entangle during film formation. The weak bonds formed by the emulsion polymer are fundamental to its dispersibility in tap water. For example, unlike the prior art, large amounts of extremely hydrophilic monomers such as acrylic acid are not needed to provide a water-dispersible composition. Less hydrophilic monomers such as methacrylic acid may be used which generally accommodates emulsion polymerization techniques better. Accordingly, in his technical opinion the compositions described in the '317 *Komatsu* reference are not remotely suggestive of the latex polymer binder used in the non-woven material of the invention.
9. The use of emulsion polymerized resins (referred to simply as "emulsion polymers," or the like, in the pending application) has significant advantages over solution polymerized resins. For example, emulsion polymerized resins exhibit a much lower viscosity for a given solids content. Example 1 of *Komatsu* reports a solids content of 17.9 % and a viscosity of 500 cps, whereas the colloid stabilized emulsion resin of Example 11 in the '129 *Eknoian* patent has a solids content of 29.7 % and only has a viscosity of 136 cps. Moreover, because the viscosities are so low, the emulsion product can be prepared and shipped at an extremely high solids content, which is advantageous from an economic perspective. Additionally, because the emulsion polymerization occurs in the dispersed phase of an aqueous medium, no solvent is required to dissolve the components. This is highly preferred, because there is no need to evaporate off excess organic solvents which are detrimental to the environment.
10. Regarding the enablement of the pending claims with respect to the glass transition temperature element, the Declarant reiterates from his previous *Declaration* that it

would be within the general knowledge of those skilled in the polymer arts to produce a resin with the claimed glass transition temperature of -40°C to 105°C. In this regard, see **Exhibit A** attached hereto from Ullmann's Encyclopeida of Industrial Chemistry, Sixth, Revised Edition, v. 28, pp. 13-14 at section 4.2.

4.2. Properties of the Polymers

Glass Transition Temperature. The glass transition temperatures (T_g) of polyacrylates and homopolymers of monomers that are frequently used in acrylate copolymers are summarized in Table 8. The values were determined by differential thermoanalysis on dispersion films (heating rate 20 °C/min, midpoint), the films being repeatedly heated until reproducible values were obtained. Other factors that influence the glass transition temperature (e.g., molecular mass, [103], [104]) are discussed elsewhere [105], [106].

The glass transition temperature of most copolymers can be accurately calculated with the nonlinear equation of GORDON and TAYLOR [107] given below. Other nonlinear equations are discussed elsewhere [108].

$$T_g = \frac{T_{gA}w_A + cT_{gB}w_B}{w_A + cw_B}$$

where w_A and w_B are the mass fractions of the monomers A and B, and T_{gA} and T_{gB} are the absolute glass transition temperatures of the corresponding homopolymers ($T_{gA} < T_{gB}$). The constant c takes into account the expansion coefficients of the melt and glass state of the two

homopolymers. Its value is generally between 0.5 and 2.0.

With the exception of *tert*-butyl acrylate, most acrylate homopolymers have extremely low glass transition temperatures. They are therefore too soft, too tacky, or have too high an elongation and insufficient strength for many areas of application. Polymers for specific applications can be produced by copolymerization with monomers whose homopolymers have high glass transition temperatures (e.g., styrene, acrylonitrile, methyl methacrylate, ethyl methacrylate).

As stated in **Exhibit A**, the glass transition temperature of a polymer can be accurately predicted by the monomer components, *i.e.*, by selecting various "hard" and "soft" monomers. This relationship is well known in the art. Furthermore, while the glass transition temperature of the latex polymers used in the invention will be somewhat effected by the polymeric colloid, the composition of the polymer will predominantly influence the Tg value. In any event, in his technical opinion, latex polymers having the claimed Tg could be readily produced by a person of ordinary skill in the art without undue experimentation.

11. The undersigned Declarant declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the subject application or any patent issuing thereon.

Dated 02-MAR-2007



Steven P. Pauls, Sr.

Ullmann's Encyclopedia of Industrial Chemistry

Sixth, Completely Revised Edition

Volume 28

Polyacrylates
to
Polyurethanes

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Exhibit A
A2

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Cross Reference:

Poly(Acrylic Acids)
Poly(Acrylic Acids):
Polyacrylic Fibers →
Organic 13
Polyalkenamers → R
Poly(Alkylene Glyc)
Polyoxyalkylenes
Polyamide Fibers →
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Polyformaldehyde

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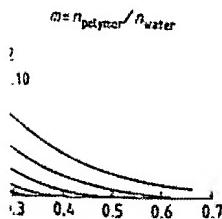
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2-Ethylhexyl acrylate	
³	not regulated
500 mg/24 h, mild	500 mg/open, mild
5660	
8460	
³	

for determining the mean particle radius of spheres theory [83]. The particle function of the light transmission important parameter is the refractive index of the polymer approximate refractive index can be calculated with the

ons $w_1 + w_2 + w_3 \dots = 1$ and indices of the homopoly-



size can also be determined by scattering (measurement

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range: 300–1500 nm). Hydrodynamic diameters can be accurately measured by *quasielastic light scattering* (QELS); measurement range: 10–3000 nm) [84–88].

The particle size distribution can be determined with the *ultracentrifuge* (measuring range: 10–20 000 nm in a gravity field of 20–200 000 g) [89]. The particles must be spherical.

The surface areas of latex (dispersion) particles are determined by *soap titration*, from which the particle size can be calculated [90–93]. Soap titration is simple and quick to perform, and yields reasonable values with homologous polymer dispersion series. Care should be taken, however, when evaluating latex surfaces of different hydrophilicity.

Viscosity. Dispersions with solids contents of 40–60 wt % exhibit a broad spectrum of rheological behavior [94], [95] that depends on the volume fraction, temperature, particle size, particle size distribution, and auxiliary system. Dispersions may be highly liquid to pasty, and may exhibit a Newtonian to viscoelastic flow behavior [96]. In dispersions structural viscosity ("shear thinning") is frequently observed; dilatancy is observed less often; thixotropy and rheopexy are rarely encountered.

Stability. Dispersions must have a certain degree of mechanical (shear) stability if coagulation is not to occur during pumping, stirring, or spraying. Mechanical stability is checked by determining the amount of coagulate after a stirring test; the stirrer, temperature, shear velocity, time, and other parameters should correspond to possible stresses occurring during use.

Thermal stability is checked by subjecting the dispersions to the anticipated service temperature conditions. A freeze-thaw test with five cycles is widely used. However, the results depend on the container size and temperature control so that the correlation between laboratory results and frost stability under relevant weathering conditions is generally unsatisfactory.

The polymer dispersion must be stable to electrolytes when diluted with hard water and when formulating products such as paints or adhesives. The stability is therefore tested by adding salts.

Polyacrylates

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Minimum Film-Forming Temperature. Whether a dispersion forms a coherent film after evaporation of the water or whether a brittle, pulverizable layer is formed depends on the glass transition temperature of the polymer, residual water content, and temperature at which the film is formed. The lowest temperature at which a coherent film can still form from the copolymer dispersion is called the minimum film-forming temperature (MFT). The MFT is measured with a metal plate that has a temperature gradient [97], [98]. The temperature at which the film begins to crack or the so-called white point, at which the turbid film starts to become clear, is measured [99].

The MFT depends on the copolymer composition, as well as the particle size and polarity of the comonomers [100–102]. It is usually comparable to the glass transition temperature, but may be substantially lower, probably because the emulsifier and water act as a plasticizer.

4.2. Properties of the Polymers

Glass Transition Temperature. The glass transition temperatures (T_g) of polyacrylates and homopolymers of monomers that are frequently used in acrylate copolymers are summarized in Table 8. The values were determined by differential thermoanalysis on dispersion films (heating rate 20 °C/min, midpoint), the films being repeatedly heated until reproducible values were obtained. Other factors that influence the glass transition temperature (e.g., molecular mass, [103], [104]) are discussed elsewhere [105], [106].

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$$T_g = \frac{T_{gA}w_A + cT_{gB}w_B}{w_A + cw_B}$$

where w_A and w_B are the mass fractions of the monomers A and B, and T_{gA} and T_{gB} are the absolute glass transition temperatures of the corresponding homopolymers ($T_{gA} < T_{gB}$). The constant c takes into account the expansion coefficients of the melt and glass state of the two

Table 8. Properties of homopolymers

Monomer	CAS registry	T_g , °C	d_{4}^{20}	n_D^{25}
	no. of polymer			
Methyl acrylate	[9003-21-8]	21	1.20	1.479
Ethyl acrylate	[9003-32-1]	-8	1.13	1.464
<i>n</i> -Propyl acrylate	[24979-83-6]	-25	1.10	1.462 ^b
<i>n</i> -Butyl acrylate	[9003-49-0]	-43	1.06	1.474
Isobutyl acrylate	[26335-74-0]	-17	1.05	1.462 ^b
<i>sec</i> -Butyl acrylate	[30247-35-6]	-6	1.06	1.462 ^b
<i>tert</i> -Butyl acrylate	[25232-27-3]	55	1.03	1.468
<i>n</i> -Hexyl acrylate	[27103-47-5]	-51	0.98	1.468 ^b
2-Ethylhexyl acrylate	[9003-77-4]	-58	0.99	1.433 ^b
Lauryl acrylate	[26246-92-4]	-17	0.99 ^c	1.468 ^b
Methyl methacrylate	[9011-14-7]	105	1.19	1.488
Ethyl methacrylate	[9003-42-3]	67	1.12	1.483
<i>n</i> -Butyl methacrylate	[9003-63-8]	32	1.06	1.483
Isobutyl methacrylate	[9011-15-8]	64	1.05	1.475
<i>tert</i> -Butyl methacrylate	[25169-00-8]	102	1.02	1.460
<i>n</i> -Hexyl methacrylate	[25087-17-6]	-14	1.01	1.479
Acrylonitrile	[25014-41-9]	105	1.18	1.519
Methacrylonitrile	[25067-61-2]	112	1.18	1.545 ^b
Styrene	[9003-53-6]	107	1.05 ^d	1.591 ^d
<i>trans</i> -1,4-Butadiene	[40022-02-4]	-107	0.97	1.518
<i>cis</i> -1,4-Butadiene	[40022-03-5]	-102	1.01	1.52
Vinyl acetate	[9003-20-7]	42	1.19	1.463
Vinyl propionate	[25035-84-1]	8	1.14	1.465 ^b
Vinyl chloride	[9002-86-2]	77 ^e	1.40	1.545
Vinyldiene chloride	[9002-85-1]	~18 ^f	1.84 ^g	1.618 ^{b,g}
Acrylic acid	[9003-01-4]	130 ^h	1.54 ^c	1.544 ^b
Methacrylic acid	[25087-26-7]	162 ^h	1.45 ^e	1.563 ^b
Acrylamide	[9003-05-8]	220 ^h	1.44 ^c	1.603 ^b
Methacrylamide	[25014-12-4]	243 ^b	1.40 ^c	1.616 ^b

^a Emulsion polymers or aqueous solution polymers, differential scanning calorimetry, 20 °C/min, midpoint. ^b From the refractive increment. ^c From the specific volume. ^d Acetone. ^e Polymerization temperature 90 °C. ^f Amorphous. ^g Crystalline. ^h After repeated heating.

homopolymers. Its value is generally between 0.5 and 2.0.

With the exception of *tert*-butyl acrylate, most acrylate homopolymers have extremely low glass transition temperatures. They are therefore too soft, too tacky, or have too high an elongation and insufficient strength for many areas of application. Polymers for specific applications can be produced by copolymerization with monomers whose homopolymers have high glass transition temperatures (e.g., styrene, acrylonitrile, methyl methacrylate, ethyl methacrylate).

Molecular Mass. See also → Plastics, Analysis, Chap. 5. The molecular mass can be calculated from the intrinsic viscosity $[\eta]$ and a knowledge of the two constants K and α :

$$[\eta] = K \cdot M^{\alpha}$$

Values for K and α in various solvents are listed in Table 9.

Molecular mass determination does not generally present any difficulties with solution polymers. With acrylate emulsion polymers, however, cross-linking of the dispersed particles may cause problems. In this case it is impossible to produce a molecular-disperse solution. Before the molecular mass is determined it must therefore be checked whether a true dilute solution exists.

Mechanical Properties of Polymer Films. The temperature dependence of mechanical moduli [112], [113] provides detailed information about the mechanical behavior and structure of polymer films. Stress-strain diagrams

Table 9. Determination of the no-

Polymer	Tetrahydrofuran ([109])	$K \times 10^3$, cm ³ /g
Poly(methyl acrylate)	10.0	C
Poly(ethyl acrylate)	8.9	C
Poly(<i>n</i> -propyl acrylate)	11.9	C
Poly(<i>n</i> -butyl acrylate)	7.4	C
Poly(isobutyl acrylate)	12.7	C
Poly(<i>tert</i> -butyl acrylate)	43.4	C
Poly(<i>n</i> -hexyl acrylate)	5.5	C
Poly(2-ethylhexyl acrylate)	11.1	C
Poly(<i>n</i> -lauryl acrylate)	27.3	C

of polymer films [114] show that the mechanical properties of the films are thus obtained at break are relatively high degree of orientation apparatus; rupturing the films, however, due to film slippage often more appropriate for a specific elongation occurs when a defined stress-strain curve of the stress-strain modulus can be accurate in tensile tests with strain.

Poly(methyl acrylate) shows a low elongation at break and a low rupture strength at room temperature. poly(ethyl acrylate) and the rupture strength of poly(ethyl acrylate) is intermediate. The elongation at break is extremely high and the rupture strength is low; the films are not easily handled. Poly(2-ethylhexyl acrylate) has an even higher elongation at break and a higher strength.

Tackiness. Poly(methyl acrylate) is very tacky at room temperature, while poly(ethyl acrylate) is only slightly tacky.

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